High-Resolution Spectroscopy of the ^{39}K Transitions at 770 nm and $^{13}C_2H_2$ Saturated Lines by a Solid-State Laser at 1.54 μm: Toward an Accurate Frequency Standard in the Optical Communication Band

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***Abstract*—A novel and compact Er-Yb: glass laser was developed with single-frequency output power > 10 mW in a wavelength range from 1531 to 1547 nm. The laser output was efficiently frequency doubled at 770.1 nm in a single-pass periodically poled lithium niobate waveguide and a second harmonic power in excess of 15 μW was achieved, with an internal conversion efficiency of $\sim 220\%\ W^{-1}$. This frequency-doubled beam was used for high-resolution saturated spectroscopy of the $^{39}K\ 4S_{1/2}$-$4P_{1/2}$ absorption line. The best resolved crossover peak showed 16% line contrast and 10-MHz linewidth. In a second experiment, working at the fundamental frequency, saturation spectroscopy of $^{13}C_2H_2$ was achieved using a low-pressure gas cell inside a build-up cavity. The measured absorption dip has 3% line contrast and 1.2 MHz linewidth. Both the atomic and molecular saturated lines are very well suited for frequency locking in order to establish accurate frequency standards in the optical communication band.**

***Index Terms*—1.5 μm, acetylene, erbium, frequency stability, potassium, solid (state) laser, spectroscopy.**

I. INTRODUCTION

FOR many applications, such as optical communication systems [1], optical fiber sensors [2], high-resolution spectroscopy, and metrology, a frequency-stabilized laser source in the 1.5-μm spectral region is of great interest. Several authors have indeed reported absolute frequency stabilization of 1.5-μm semiconductor lasers against atomic [3]–[5] or molecular [6]–[8] absorption lines. Diode lasers, however, even when used in an external-cavity configuration, exhibit relatively poor spectral qualities as compared to solid-state lasers. Diode-pumped bulk Er–Yb lasers oscillating at 1.5 μm[9], on the other hand, are very attractive sources for amplitude and frequency stabilization [10] due to their intrinsic stability, wide wavelength tunability [11], and excellent beam quality. High-resolution spectroscopy of both atomic and molecular transitions is here discussed toward the realization of a frequency standard in the 1.5-μm optical band. In this work, we first report on a novel Er–Yb: glass microlaser [12], with significantly improved stability and reliability performance, particularly suitable for absolute frequency stabilization experiments. The main novelties are related to a very compact design of the laser cavity, with a massive and hermetically sealed-off laser structure and to the use of a fiber-coupled diode laser pumping. These innovations resulted in a decrease of acoustic vibrations of the cavity and in higher stability of the optical alignment between pump radiation and lasing medium.

In a first set of experiments, high-efficiency single-pass frequency doubling of the 1.5-μm beam was obtained in a periodically poled lithium niobate (PPLN) quasi- phase-matched waveguide [13]. This second-harmonic signal was used for high-resolution spectroscopy of the D_1 line of ^{39}K at 770.11 nm in a low-pressure atomic sample. The narrow linewidth of the laser source enabled clear determination of all the saturation dips and crossover lines corresponding to the hyperfine structure of the $4S_{1/2}$-$4P_{1/2}$ transition in this atom; these lines are very narrow absolute frequency references and can be used for an effective stabilization of the laser frequency. The frequency stability eventually achieved at 770 nm would be transferred to the fundamental frequency, thus obtaining an absolute frequency standard at 1.54-μm wavelength.

In a second set of experiments, the fundamental frequency of an Er-Yb laser at ~ 1.54 μm wavelength was used, for the first time to our knowledge, to perform saturation spectroscopy of the acetylene molecule. Even in this case, an extremely narrow saturation dip was observed in the Doppler profile of the P(16) line of $^{13}C_2H_2$. The clear advantage of working at the fundamental frequency, i.e., without a frequency doubler,

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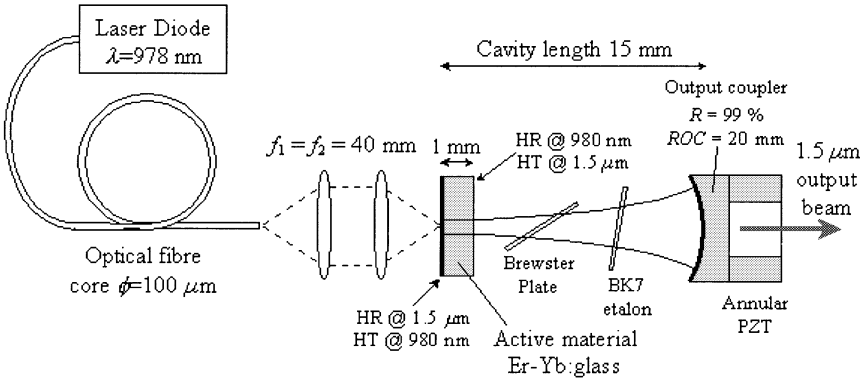

Fig. 1. Schematic diagram of the diode-pumped Er-Yb: glass laser.

is somehow attenuated by the need for an external build-up cavity used to increase the beam intensity in order to saturate the 1.54-μm transition. In this case, even to perform saturation spectroscopy, a first frequency-locking loop is needed and to go on with the absolute frequency stabilization, two frequency-locking loops must be working at the same time, thus increasing complexity of the system.

II. THE ER–YB LASER

A novel diode-pumped Er–Yb: glass microlaser was developed, with a very compact and noise insensitive cavity design. In this way we obtained a consistent reduction of the frequency noise, mainly due to mechanical vibrations, for the free-running laser. The active material was pumped by means of a fiber-coupled diode laser, further improving the ruggedness and compactness of the laser device. A single-frequency output power up to 20 mW was obtained in a diffraction-limited TEM$_{00}$ mode. The use of a phosphate glass host for the active Er^{3+} ions allows for a very broad wavelength tunability in the 1.54 μm spectral region [11]; in particular, the oscillator is continuously tunable, in single-frequency operation, from 1531 to 1547 nm.

A schematic drawing of the Er–Yb laser is shown in Fig. 1; the optical cavity consists of a plano-spherical resonator of $\sim$ 15.5 mm optical length. A dichroic coating on the active medium (power reflectivity $> 99.9\%$ at 1.5 μm and power transmission $> 96\%$ at 978 nm) acts as the input mirror, whereas the output coupler is a spherical mirror with 20 mm radius of curvature (*ROC*) and 1% transmission at the laser wavelength. The active medium consists of a 1 mm thick and 3×3 mm^2 wide square platelet, made of a special phosphate glass (Kigre Inc., QX/Er) doped with Er^{3+} at 0.73 $\times 10^{20}$ ions/cm^3 and Yb^{3+} at 1.9×10^{21} ions/cm^3. A 1 W laser diode emitting at 978-nm wavelength, coupled to a 100-μm core diameter and 0.13-NA optical fiber (Opto Power Corporation, H01-A001-FC/100), provides for the pump beam, which is focused in the active medium by a 1:1 image of the beam waist at the output of the fiber. The second facet of the active medium, inside the laser cavity, is antireflection (*AR*) coated at the laser wavelength ($R < 0.2\%$ at 1.5 μm) and high-reflectivity (*HR*) coated at the pump wavelength ($R > 94\%$ at 978 nm). The backreflection of the unabsorbed pump power results in a double-pass pumping, allowing for a higher and more uniform population inversion along the Er-Yb phosphate glass. In these quasi-three-level lasers, indeed, unpumped regions within the active medium would result in reabsorption losses. Furthermore, a more uniform pumping profile increases the pump power damage threshold of the active material, which, in our case, is a severe problem, because of the rather poor thermal conductivity ($\sim$ 0.85 Wm^{-1}K^{-1}) of the phosphate glass. However, it was observed that, even with double-pass pumping, pump powers in excess of $\sim$ 500 mW could produce a permanent damage (crack) in the laser platelet as a consequence of excessive local heating and thermal gradient within the glass host.

During our experiment, the threshold pump power for this laser was 95 mW and the slope efficiency was 6%, in single-frequency operation and on a fundamental TEM$_{00}$ transverse mode (see Fig. 2). The maximum laser output power was kept below 14 mW in order to work within a very safe pump power level of 330 mW. An upper limit to the pump power in our working conditions lies in the 400–600-mW range, depending on the residual stress and tensions left in the phosphate glass after the cutting and polishing processes. New strengthened glasses, already produced by Kigre Inc., should enable increasing the damage threshold by a factor of three, thus allowing for laser outputs of the order of 50 mW. Actually, single-frequency output powers of slightly more than 50 mW have already been demonstrated even using the unstrengthened glasses. Unfortunately, a permanent damage in this glass is very likely to occur under these high pump powers.

Single longitudinal mode selection and coarse spectral tuning were obtained by inserting within the laser cavity a 50-μm glass etalon, coated for a 10% reflectivity at the laser wavelength. By tilting the etalon, a continuous tuning from 1531 to 1547 nm was achieved, within the etalon-free spectral range of 16 nm. The typical oscillation linewidth was below 50 kHz in 1 ms. An annular piezoelectric transducer (PZT), glued to the output mirror, allowed for a fine tuning of the cavity length and thus of the laser frequency. The PZT we used (Pickelmann, HPSt-500/10-5/15) provided for the $\lambda/2$ excursion with a control signal of only 32 V. The mode-hop-free frequency tuning range was 9.7 GHz with a tuning coefficient of 290 MHz/V. This frequency span

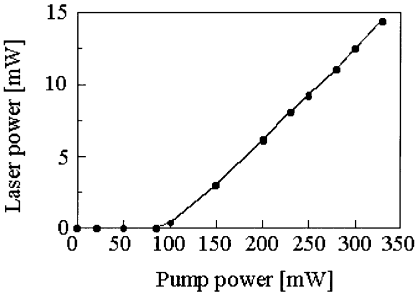

Fig. 2. Single-frequency output power at 1.5 μm as a function of the incident pump power.

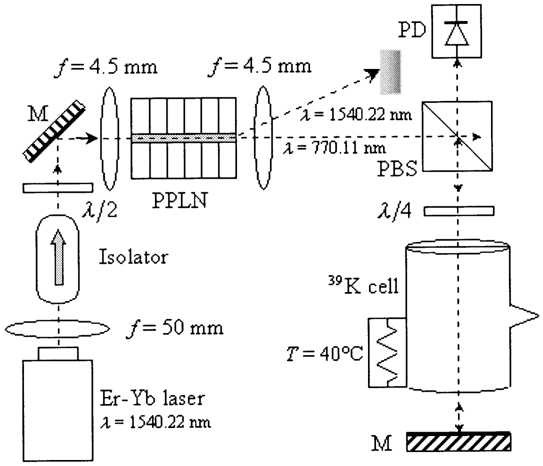

Fig. 3. Schematics of the experimental setup for frequency doubling of the Er-Yb: glass laser and high-resolution spectroscopy of the ^{39}K line; M: Mirror; PD: silicon photo detector; PBS: polarizing beam splitter.

was wide enough for performing high-resolution laser spectroscopy of different molecular/atomic transitions falling in this spectral region.

III. FREQUENCY DOUBLING

The 1.5-μm output beam from the Er-Yb laser was coupled by a microscope lens (focal length 4.5 mm) into a 30-mm long, 2.5×4 μm-wide annealed proton exchange channel waveguide made in a PPLN crystal as described in [14]. An optical isolator (Optics For Research, IOT-4-1550, > 60 dB isolation) was used to avoid feedback to the laser cavity from the uncoated input face of the crystal (see Fig. 3). Quasi-phase-matched doubling at a wavelength of 1540.2 nm was achieved by keeping the waveguide doubler at a temperature of 62 ± 1 °C. The measured second harmonic power was 17 μW with a 1.54-μm input power of 8 mW and the external conversion efficiency was therefore $\sim 26\%$ W^{-1}. Taking into account the losses owing to the coupling to the waveguide (approximately 50%), the Fresnel reflections of the input (14%) and output (20%) beams and the losses in the waveguide ($\sim 20\%$), the internal second harmonic efficiency was $\sim 220\%$ W^{-1}. This conversion efficiency was two to three orders of magnitude higher than the efficiency obtained in bulk frequency doublers. However, with higher input powers, which can be potentially generated by Er-Yb lasers, bulk frequency doublers may also be used. Assuming, e.g., a pump power of 50 mW and a bulk conversion efficiency of 1% W^{-1} (which can be obtained with a 1 cm long PPLN crystal under optimal focusing conditions), the generated second harmonic power is 25 μW. Bulk doublers are often preferred because of the simpler production and less critical alignment.

IV. HIGH-RESOLUTION SPECTROSCOPY OF ^{39}K

Atomic absorptions are typically preferable with respect to molecular ones in order to achieve narrow saturated transitions with comparatively low saturation intensity; several groups have used the second harmonic of 1.5 μm semiconductor lasers to probe the saturated Rb [15]–[17] or K [5], [18] lines in the spectral range from 770 to 780 nm, or to frequency lock the laser to these lines. In particular, the $4S_{1/2} \rightarrow 4P_{1/2}$ ^{39}K transition at 770.11 nm has a saturation intensity lower than 100 μW/mm^2 [5], whereas, to give a comparison, the saturation intensity of the $^{13}C_2H_2$ transition in the 1.55-μm band, even at a rather low gas pressure of 4 Pa, is as high as 6 W/mm^2 [8]. We used the ^{39}K atomic reference, because the corresponding fundamental wavelength (1540.22 nm) could be obtained with reasonably high power ($\sim$ 8 mW) with our Er-Yb microlaser. Both ^{39}K and ^{41}K isotopes were contained in a cell in their natural mixture of 93.1% and 6.9%; the vapor pressure was increased up to $\sim$ 13 μPa heating the cell to 40 °C. In our case, we worked with the $4S_{1/2}$-$4P_{1/2}$ transition of potassium (see Fig. 4). This provides for nine different transitions between its hyperfine level structure where each level is split into two sublevels, depending on the combination of the nuclear spin, $I = 3/2$, and the total angular momentum of the electron $J = 1/2$, hence leading to a total spin number $F = I \pm J = 1, 2$. The distance between the $F = 2$ and $F = 1$ sublevels in the $4P_{1/2}$ and $4S_{1/2}$ is 55.6 and 462 MHz, respectively, as indicated in Fig. 4.

The second harmonic beam was sent to the K cell as a nearly collimated beam with $\sim$ 200 μm diameter and $\sim$ 17 μW power level. Sub-Doppler spectroscopy of the ^{39}K line was then performed by a standard pump and probe technique. During the spectroscopic measurements, first of all, we set the laser wavelength in proximity of the absorption line by tilting the intracavity etalon and then a ramp voltage was applied to the PZT in order to sweep the laser frequency through the atomic resonance. The optical power transmitted through the K cell is monitored by a photodetector (see Fig. 3) and a typical transmission curve of the ^{39}K line is reported in Fig. 5: the full width at half maximum (FWHM) is $\sim$ 1.7 GHz. Several absorption dips appeared within the Doppler profile (see Fig. 5), corresponding to different transitions between the four hyperfine structure levels of the $4S_{1/2}$-$4P_{1/2}$ transition and to crossover frequencies (see Fig. 4). The best resolved line in Fig. 5 is the absorption peak corresponding to the crossover line labeled (5), with a linewidth of $\sim$ 10.6 MHz and a line contrast (with the available laser beam intensity of $\sim$ 1 mW/mm^2) resulting in 16% with respect to the Doppler-broadened line. This line could be therefore the best absolute reference for a frequency stabilization of our laser. In addition, we measured the first and third derivative of the signal corresponding to this crossover line by wavelength-modulation spectroscopy and synchronous detection. A frequency modulation was superimposed to the laser frequency by applying to the PZT a sinusoidal signal with

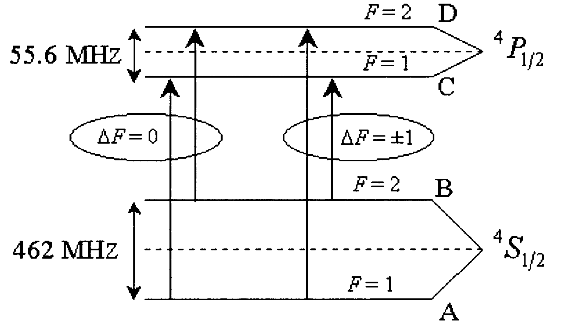

Fig. 4. Hyperfine structure levels of the $4S_{1/2}$-$4P_{1/2}$ transition of potassium.

frequency $f_m = 423$ Hz and amplitude 5 mV, providing for a peak-to-peak frequency deviation of 2.9 MHz of the frequency doubled laser beam. In order to retrieve the first and third derivative signals, the output voltage from the photodetector was sent to a lock-in amplifier, set with a sensitivity of 500 μV and a time constant of 3 ms. The obtained first derivative, amplified by a factor of 50 V/V, is shown in Fig. 6. This signal shows an odd symmetry with respect to the resonant frequency and, therefore, can be used to lock the laser frequency to the central frequency of the crossover line. The measured linewidth of the crossover line was $\sim$ 10.6 MHz and the slope of the derivative signal at the output of the lock-in amplifier, including the gain factor of 50, was 2.16 μV/Hz with a S/N ratio of 50 dB in a 53-Hz bandwidth. The third derivative signal provided improved suppression of the linear absorption background, but the signal-to-noise ratio was substantially lower in this case. Therefore, the first derivative is preferable, despite the nonzero bias, as the output discriminating signal for a frequency stabilization loop.

V. SATURATION SPECTROSCOPY OF $^{13}C_2H_2$

Acetylene ($\nu_1 + \nu_3$) rovibrational absorption bands at 1.54 μm wavelength provide for very interesting absolute frequency references in this near-infrared spectral region. The acetylene molecule, in fact, shows 65 clean and well-resolved absorptions from 1519 nm to 1553 nm (considering transitions with $> 10^{-22}$ cm/mol line strength). In addition, this molecule is stable, not toxic and does not have a permanent electric dipole moment. Working with gas samples at room temperature, the Doppler broadening of these absorption lines keeps the reference width at more than $\sim$ 500 MHz [19], whereas using saturation line-dips at low gas pressure, a natural linewidth of $\sim$ 1 MHz [6] can be obtained at $\nu_0 \cong 200$ THz with a quality factor $Q = \nu_0/\Delta\nu \sim 2 \times 10^8$ for the molecular reference. Using saturated absorptions of the $^{13}C_2H_2$ molecule as a frequency reference, other research groups [6], [8] achieved a 10^{-12} level for the Allan deviation with integration times > 1 s, as measured with beat note experiments between two independently stabilized semiconductor-laser sources.

In our experiments, in order to obtain a saturated absorption signal, we used a Brewster-window quartz cell filled with $^{13}C_2H_2$ at a pressure of 4 Pa placed inside a Fabry–Perot cavity [12]. This is a passive plano-spherical resonator, 30 cm long, with a 2.5-MHz linewidth and a finesse of 200. The measured finesse, when the laser frequency was on the acetylene resonance, is decreased to $\sim$ 140 (observed linewidth of 3.5 MHz). To keep the resonance condition between the laser frequency and the Fabry–Perot cavity, a standard Pound–Drever–Hall locking setup was adopted. Using a modulation frequency of 25-MHz and a phase modulation index of $\sim$ 0.2, the laser frequency was locked to the resonance of the cavity by means of an electronic servo system closed on the laser PZT with a control bandwidth of $\sim$ 1 kHz (limited by the PZT mechanical resonance). The optical power incident on the Fabry–Perot cavity was 9 mW and about 75% of this power was coupled into the fundamental mode of the resonator.

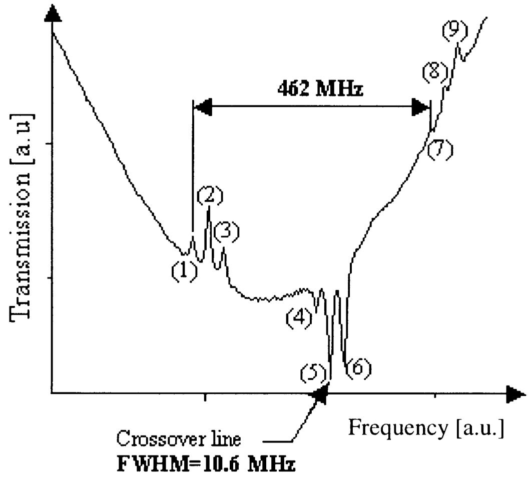

Fig. 5. Saturation spectroscopy of the ^{39}K sample as obtained by scanning the Er-Yb laser frequency through the Doppler-broadened resonance.

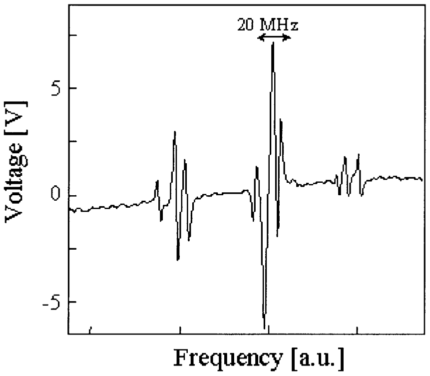

Fig. 6. First derivative voltage signal as obtained at the lock-in output after amplification of 50 V/V.

With the laser frequency locked to the cavity resonance, by scanning the length of the passive resonator, it was possible to observe the saturated absorption profile of the $^{13}C_2H_2$ rovibrational line by means of the cavity transmission signal. Several rovibrational lines of $^{13}C_2H_2$ [from P(10) to P(16)] were observed, and an example of the recording of the transmission profile for the P(16) line, at a wavelength of 1542.384 nm, is shown in Fig. 7. In this spectral profile, a Lamb-dip of relative contrast of 3% with respect to the linear absorption (linewidth of $\sim$ 500 MHz) appears right at the line center with a full width of $\sim$ 1 MHz [6], [20]. In our setup, taking into account the cavity finesse, the coupling factor, the beam profile inside the cavity, and the phase modulation index, the saturating intensity inside the cavity was evaluated to be in the order of 3.5 W/mm^2.

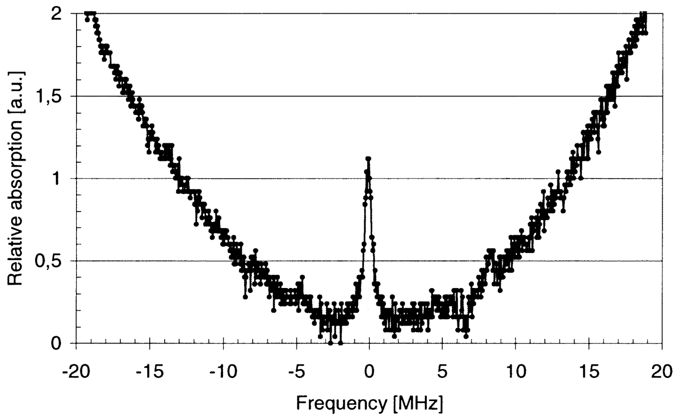

Fig. 7. Saturation spectroscopy of the $^{13}C_2H_2$ P(16) line at $\lambda = 1542.384$ nm. The saturation line contrast is $\sim 3\%$ and its linewidth is 1.2 MHz.

In addition, we measured the first derivative of the signal corresponding to the saturated line of Fig. 7 using wavelength-modulation spectroscopy and synchronous detection. A sinusoidal frequency modulation was superimposed to the laser frequency, providing for an optical frequency deviation of 2 MHz. This was achieved by applying to the PZT of the Fabry–Perot cavity a voltage signal at 350 Hz with amplitude 250 mV. In order to retrieve the first derivative signal, the output voltage from the photodiode detecting the Fabry–Perot transmission was sent to a lock-in amplifier, set with a sensitivity of 50 mV and a time constant of 30 ms. The obtained first derivative is shown in Fig. 8 and the slope of this signal was $\sim$ 1 V/MHz with a S/N of 40 dB in a 3-Hz bandwidth.

VI. CONCLUSION

A novel and very compact single-frequency Er–Yb bulk microlaser at 1.54 μm was developed and used for high-resolution spectroscopy experiments. The TEM_{00} output beam was frequency doubled with high efficiency in a periodically poled lithium niobate waveguide crystal. Up to 17 μW of 770-nm radiation were obtained starting from $\sim$ 8 mW of fundamental power. This second harmonic radiation was successfully used to achieve high-resolution saturation spectroscopy of the ^{39}K D_1 line, allowing for clear identification of the four different transitions and of the crossover lines between the corresponding hyperfine sublevels. By means of wavelength-modulation spectroscopy of the crossover line showing the best contrast, we measured the first and third derivative signals, which can be used for an effective frequency locking of the laser. In other experiments, by using a high-finesse Fabry–Perot resonator containing a low-pressure $^{13}C_2H_2$ Brewster gas cell, Doppler-free spectroscopy of the P(16) line at 1542.384 nm wavelength was achieved. Due to the excellent spatial and spectral properties of the Er–Yb laser, a very efficient mode coupling to the external resonator was achieved with $\sim$ 10 mW TEM_{00} optical power. Doppler-free absorption could be easily observed and a saturation dip with 3% line contrast and 1.2-MHz linewidth was measured.

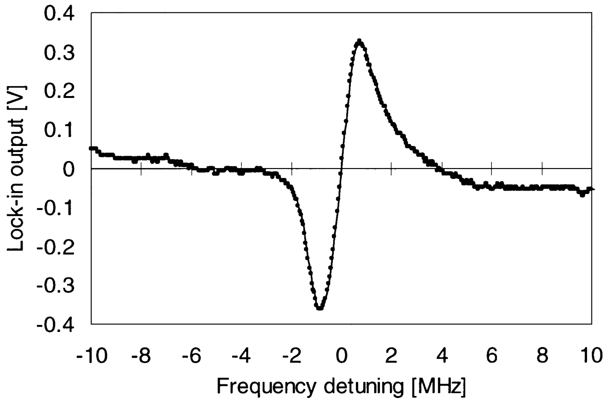

Fig. 8. First derivative of the saturated absorption dip in line P(16) of $^{13}C_2H_2$.

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R. Klein, photograph and biography not available at the time of publication.

M. A. Arbore, photograph and biography not available at the time of publication.

M. M. Fejer, photograph and biography not available at the time of publication.

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